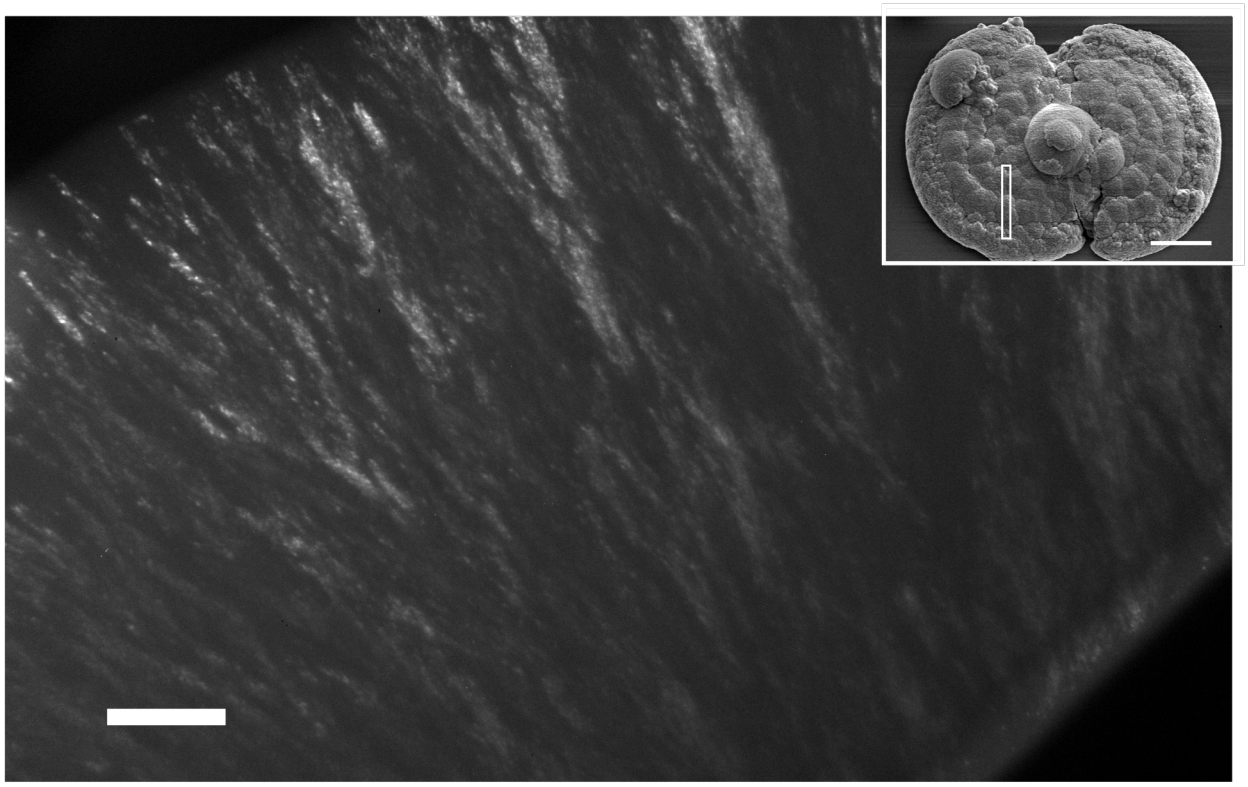

Fig S1. Cross-section of a botryoidal calcite nanoparticle aggregate (inset) that transforms from ACC under conditions with high Mg/Ca levels. TEM dark field image of FIB liftout shows high contrast (bright) regions that indicate nano-crystalline regions with similar orientations. Scale bar = 500 nm. Inset: SEM of botyroidal calcite aggregate with rectangular box that denotes the section of the sample where the FIB liftout was taken. Scale bar = 10 microns.

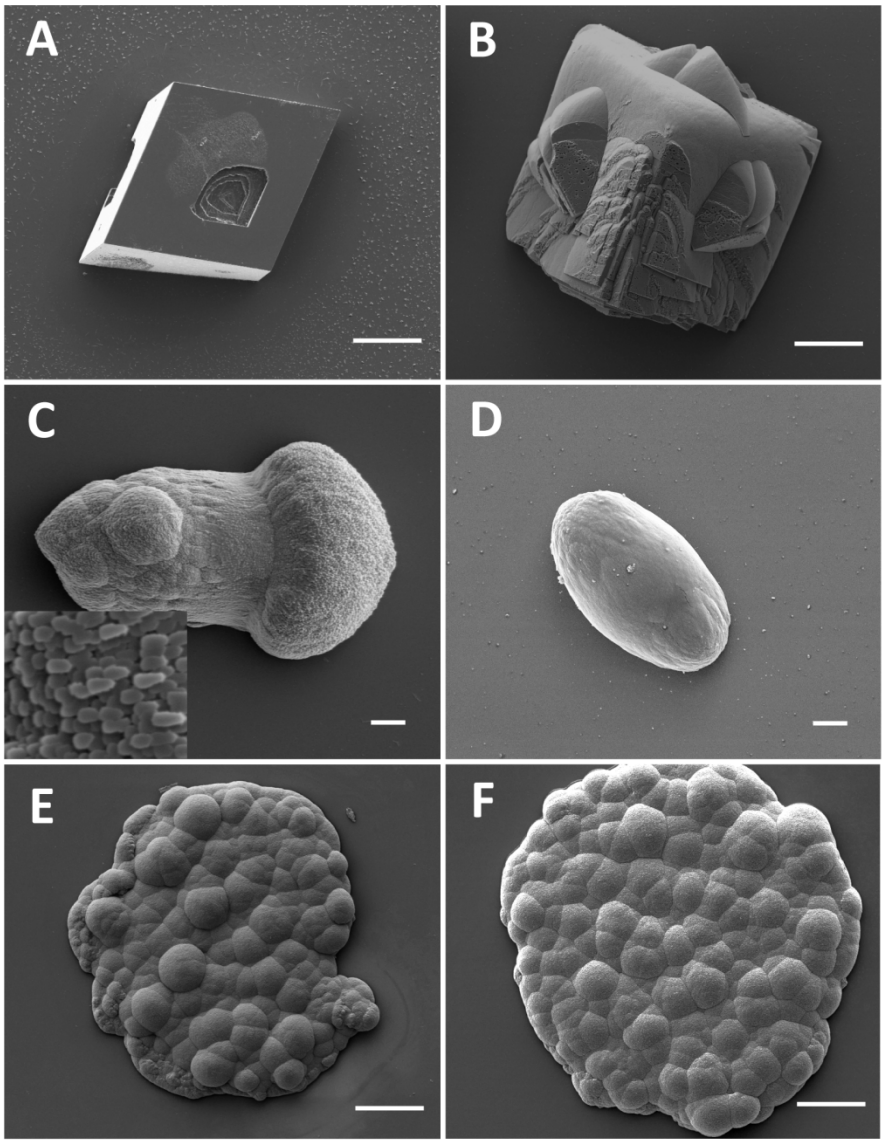

Fig. S2 The textures and compositions of calcites that form from classical and the ACC-to-crystal pathway are unaffected by sulfate or the aspartate monomer compared to controls (**Fig 1**). Mineralization in the absence of Mg (Mg/Ca = 0) produces rhombic crystallites typical of classic step growth: (**A**) 0.05 M Asp; and **(B)** 0.05 M sulfate solutions. At initial Mg/Ca = 2.0, morphology shifts to elongated mesocrystalline assemblages of crystallites: (**C**) 0.05 M Asp; and (**D**) 0.05 M sulfate. At initial Mg/Ca ratios = 5.0 and higher, ACC transforms into low-relief aggregates of discrete calcite nanoparticles: (**E**) 0.05 M Asp; and (**F**) 0.05 M sulfate solutions. All scale bars = 20 microns.

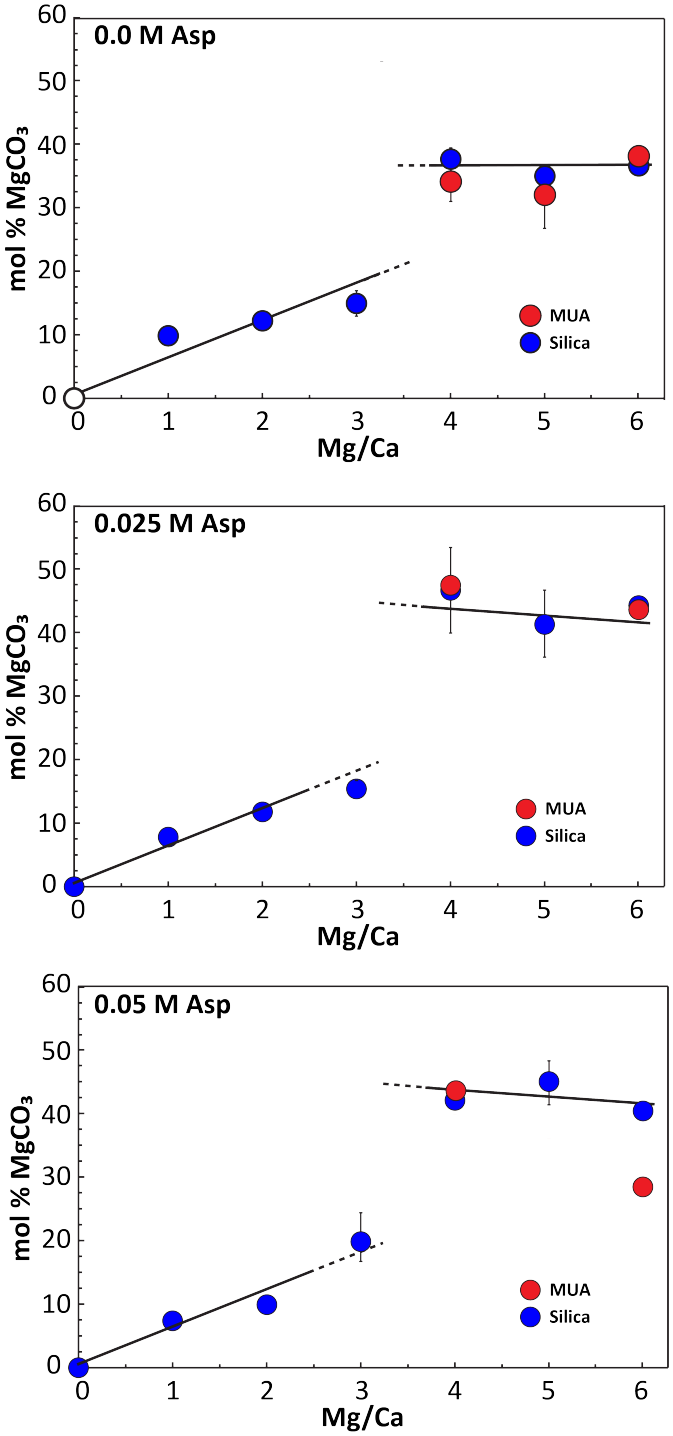

Fig. S3 Calcites that transform from ACC onto carboxylated (MUA) surface assembled monolayers show similar compositions to those measured on silica glass surfaces for the control and Asp-bearing experiments.